

The University of Chicago

- I. The Separation of Mercury into Isotopes in a
Large Apparatus
- II. A Graphical Study of the Stability Relations of
Atom Nuclei

A DISSERTATION

SUBMITTED TO THE FACULTY OF THE OGDEN GRADUATE
SCHOOL OF SCIENCE IN CANDIDACY FOR THE
DEGREE OF DOCTOR OF PHILOSOPHY

DEPARTMENT OF CHEMISTRY

BY

SAMUEL LEO MADORSKY

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THE SEPARATION OF MERCURY INTO ISOTOPES IN A LARGE APPARATUS¹

Introduction

The object of the work of the present paper was to demonstrate the efficiency of a moderately large apparatus for the separation of the isotopes of mercury by vaporization in a vacuum. This apparatus is of a design such that it may easily be constructed as a part of a large multiple unit apparatus. It has been shown by Mulliken and Harkins² that the low pressure vaporization is essentially a diffusion through the surface of the liquid, which acts as a surface of molecular thickness, with apertures of the dimensions of a molecule. While the separation coefficient of mercury (0.0060) is considerably smaller than that of chlorine as hydrogen chloride (0.00950), the apparatus for the separation of the former may be made much more simple than that needed for the latter, so that the simplicity of the apparatus more than makes up for the smallness of the coefficient, provided a certain change in atomic weight is desired. However, for a certain change in the relative density—which is probably more important, if the object in view is to demonstrate differences in properties, such as the spectra, of the isotopes obtained—the separation coefficient of hydrogen chloride is nearly 9 times as great as that of mercury, so it is very difficult to overcome the theoretical advantage of the former by the use of any form of apparatus.

The advantage is, however, very greatly on the side of mercury when convenience of manipulation is considered, but is not inherent in the vaporization method, since Dr. Mulliken, working in this Laboratory, has secured an even more rapid separation by utilizing diffusion through a thin membrane, such as filter paper. The increase in the rate of separation was in this case due to the fact that it is easy to combine an efficiency of nearly 100% in the diffusion with a somewhat smaller efficiency in the vaporization utilized to obtain the vapor for diffusion.

A Large Apparatus for the Separation of Mercury into Isotopes by Vaporization

The apparatus used for the separation of the isotopes of mercury by Brönsted and Hevesy³ is very simple, and consists merely of a Dewar

¹ Reprinted from a paper by W. D. Harkins and S. L. Madorsky, *J. Am. Chem. Soc.*, Vol. XLV, No. III, March, 1923.

² Mulliken and Harkins, *J. Am. Chem. Soc.*, **44**, 37–65 (1922).

³ Brönsted and Hevesy, *Phil. Mag.*, **43**, 31 (1922). These writers have failed to refer to the much earlier work on the separation of isotopes published from this Laboratory.

bulb, the outer jacket of which is used to contain the mercury, while the inner bulb is filled with liquid air, which acts as a cooling agent. Because the mercury is frozen by the liquid air it is difficult to collect the diffusate in several fractions. The difficulty was obviated by Mulliken and Harkins, who used an apparatus of an entirely different design, with ice as the cooling agent. This has been used almost entirely to study the efficiency of the process as related to pressure, speed of vaporization and form of apparatus, and no attempt has been made to obtain a separation of any considerable magnitude. It seemed advisable, therefore, to design a larger apparatus which would allow of the use of a considerable amount of material at one time. This was done early in 1921, and the first model was made of moderate dimensions in order that the efficiency of the process might be tested before the construction of a larger apparatus was undertaken. The general principle used is very similar to that made use of by Mullikens and Harkins in that the molecules which shoot out from the evaporating surface strike a roof, which has a slope such that the liquid mercury will adhere and run down the surface. The angle of the slope used is 45° .

The apparatus, Fig. 1, was cut from 2 large pieces of forged steel. Cast steel was not used since it is more likely to leak. The mercury is vaporized at a pressure between 0.001 and 0.0001 mm. from an annular steel trough D which holds 2600 g. of mercury. This amount could be easily increased in the same apparatus by the use of a wider trough. The trough is heated by an iron-encased calorized Nichrome wire F, insulated by magnesium oxide. This should be buried in a cast-iron jacket in order to give good contact with the trough, or it can be put inside the trough which affords better conduction of the heat, but does not give such free evaporation when the sample becomes very small. The coil may be cast into the base of the trough itself. The 4 supports for the heating wire are made from glass rods, though a poorer conductor of heat such as zirconium oxide would have been even better. The electric current is carried to the heating plate by 2 platinum wires, i, sealed in through the walls of the glass tube G which leads to the vacuum pumps. The wire is insulated from the inverted steel cone by 2 glass tubes, l. The joint, n, between the glass tube and the steel is long and very carefully ground, there is a second ground joint between the steel roof B and the inverted cone C. This is sealed by mercury, h, held in place by a thin hoop of steel, held on by friction. The cooling cylinder A is made of brass. It does not need to be as deep as the dimension given (36 cm.), but this depth is advisable in order to give a sufficient capacity for ice. The only materials, other than mercury, exposed on the inside of the apparatus are glass, steel and platinum, and the surface of the last is very small.

Operation of the Apparatus

The trough of the thoroughly cleaned apparatus is filled with 190 cc. of mercury which has been highly purified, and the roof B is carefully lowered. Mercury is run in at h to make the joint tight, and the vacuum pumps are then started. These consist of a Hyvac oil pump, and a large and very rapid Knipp mercury condensation pump. The current is then turned on, and as the heating plate rises in temperature considerable quantities of gas are evolved. The lower extremity of the glass tube G is sealed off with a long point, and in this the lightest fraction of the mercury begins to condense. If the pressure in the apparatus is sufficiently low, this has an atomic weight 0.006 unit

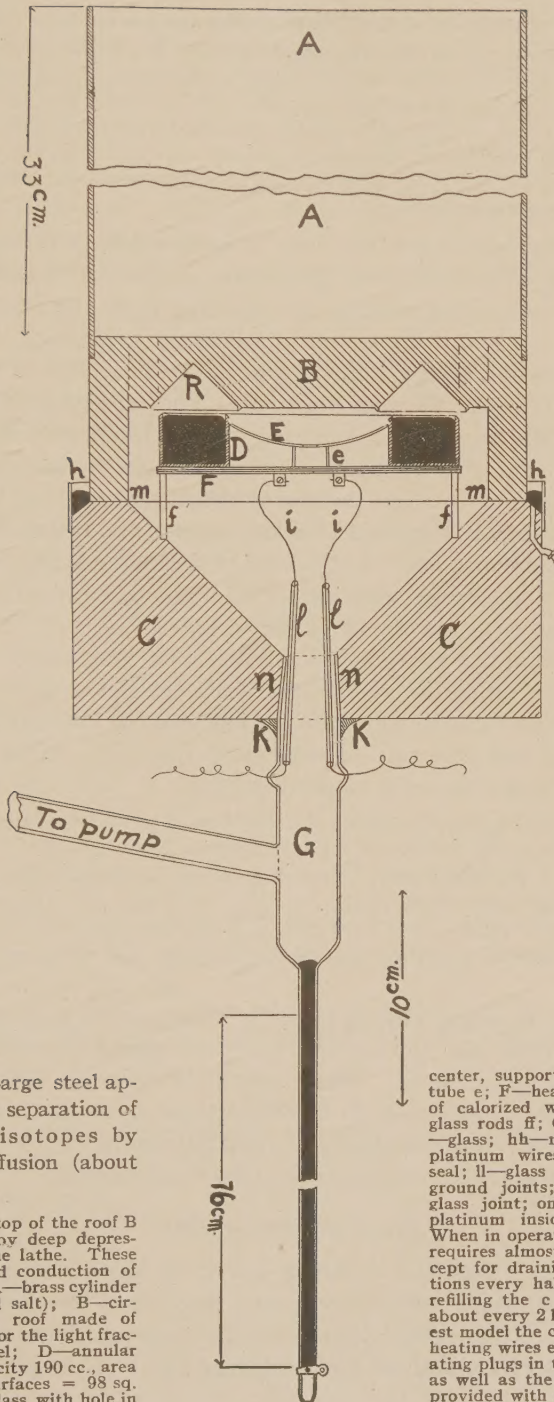


Fig. 1.—Large steel apparatus for the separation of mercury into isotopes by evaporative diffusion (about $1\frac{1}{2}$ actual size).

Note: The top of the roof B was actually cut by deep depressions made on the lathe. These give a more rapid conduction of heat to the ice. A—brass cylinder for ice (or ice and salt); B—circular condensing roof made of steel; C—drain for the light fraction made of steel; D—annular steel trough, capacity 190 cc., area of evaporation surfaces = 98 sq. cm.; E—watch glass with hole in

center, supported on short glass tube e; F—heating element made of calorized wire, supported on glass rods f; G—collecting tube—glass; hh—mercury seal; ii—platinum wires; kk—sealing wax seal; ll—glass sleeves; mm—ground joints; nn—ground steel glass joint; only steel, glass and platinum inside the apparatus. When in operation the apparatus requires almost no attention except for draining the light fractions every half hour or so, and refilling the cylinder with ice about every 2 hours. In the newest model the connections ll to the heating wires enter through insulating plugs in the base and these, as well as the glass tube G, are provided with mercury seals.

lower than ordinary mercury, so it is lighter by 30 parts per million. This is 60 times the detectable difference since the pycnometer described by Mulliken and Harkins gives the density to 1 part in 2 million or better when only ordinary precautions are used. The lower part of this tube is a capillary in order that its volume may be small, and the mercury is run out against the pressure of the atmosphere after its height has risen above 760 mm. If small fractions are desired the mercury may be run into a vacuum flask or bottle at the lower end of G, but with too small fractions the work of determining the density becomes considerable.

In the experimental work the rate of evaporation was kept at 15 to 20 cc. per hour, or 0.15 to 0.2 cc. per square centimeter of surface. Thus a run in which all of the mercury is vaporized from the trough takes at least $9\frac{1}{2}$ hours, provided the trough is full at the beginning.

The molecules of mercury which escape from the surface of the mercury in the trough D strike the roof above R. The temperature of this roof is kept low by the ice water in A, so very few molecules re-evaporate after condensation. The greatest distance which a molecule moving vertically upward has to travel between the top of the full trough and the peak of the roof, is less than 2.5 cm. and near the edges of the trough this distance diminishes to less than 1 cm. As the level of the mercury falls, the distance traveled by the molecules increases, and becomes considerable when it is nearly empty, since the depth of the trough is 2.40 cm. Furthermore, as the trough empties the molecules have more and more opportunity for condensation on and re-evaporation from the hot walls of the trough. These two factors serve to make the efficiency of the apparatus somewhat less toward the end of a run, or with small quantities of mercury, than at the beginning of a run with large quantities.

In order to obviate this evil a small conical roof has been cut from below in the center near B. A small circular dish of any capacity between 25 and 200 g. of mercury can be placed underneath this and heated by an independent hot plate. This is to be used when the samples become very small, but thus far it has not been put into operation since the glass apparatus of Mulliken and Harkins has been used for small samples. For this reason, and for the sake of simplicity, this unit is not shown in the drawing.

The mercury which collects on the surface of the steel gathers into drops, and these run down the 45° incline to a sharp edge which determines exactly where they shall drop. Those which fall from the outer edge drop directly into the conical hole in C, while those from the inner edge are collected by an inverted watch glass E, and fall through a hole at its center e, thus falling where there are no wires to be encountered.

Fractionation Scheme

Ten runs, I to V and XVI to XX of 190 cc. or 2600 g. each, using 26 kg. of mercury in all, were made on very pure mercury of the ordinary atomic weight. Each of these was collected in 8 fractions, as outlined in Table I. The process may be visualized by consulting Fig. 1 of the paper by Mulli-

ken and Harkins.⁴ The decrease of the atomic weight ($-\Delta M$) to be expected on the basis of the theory developed in this paper was calculated for the first fraction from the equation

$$-\Delta M = E.B \frac{\ln C}{C-1} \quad (1)$$

where E is the fractional efficiency, B is the separation coefficient, and $C = (N_0/N)$ is the cut. N_0 is the initial number of moles, and N is the number of moles in the residue.

For Fractions 2 to 7, that is, all of the intermediate fractions

$$-\Delta M = \frac{E.B}{C_2 - C_1} (C_1 \ln C_2 - C_2 \ln C_1) \quad (2)$$

Here C_1 and C_2 are the values of the cut at the beginning and end of the fraction which is taken.

For the end fraction (8)

$$-\Delta M = E.B \ln C \quad (3)$$

TABLE I

FRACTIONATION SCHEME FOR 190CC. OR 2600G. SAMPLES OF MERCURY OF ORDINARY ATOMIC WEIGHT. (RUNS I TO V, AND XVI TO XX)

Fraction	%	Cc.	Weight G.	Cut C or $C_2 - C_1$	Change of dens. (Parts per million)		Actual	Change of at. wt.	
					Calculated 100%	90%		100%	Actual
1	20	38	520	1.25	-25.4	-22.9	-22.8	-0.005	-0.0046
2	20	38	520	1.66-1.25	-18.2	-16.4		-0.0036	
3	15	28.5	390	2.22-1.66	-10.2	-9.2		-0.0020	
4	15	28.5	390	3.33-2.22	-0.4	-0.36		-0.00008	
5	10	19	260	5-3.33	+10.9	+9.8		+0.0022	
6	10	19	260	10-5	+26.2	+23.6		+0.0052	
7	5	9.5	130	20-10	+45.9	+41.3		+0.0092	
8	5	9.5	130	20	+85.2	+76.7	$V^a = +78$	+0.0170	+0.016

^a V designates that this particular figure was obtained in the fifth run, which gave about the average efficiency of 90%. II₈, that is the 8th fraction from the second run, showed an increase of only 51 parts per million, or an efficiency of 63%. This low efficiency was due to the high pressure, 0.060 mm., which was 60 times higher than the highest pressure in the other runs.

Table II gives the fractionation scheme used on the light fractions given in Table I.

TABLE II

FRACTIONATION SCHEME FOR THE LIGHT FRACTIONS OF TABLE I. (RUNS VI TO IX AND XXVIII TO XXXIII)

Fraction	%	Wt. G.	Cut C or $C_2 - C_1$	Change of density		Actual	Density Actual	Δ (At. wt. increase)
				Calculated 100%	90%			
1	25	650	1.33	-24.7	-22.2	-22.2	-44.4	-0.0089
2	25	650	2-1.33	-14.8	-13.3	..	..	
3	25	650	4-2	0	0	..	..	
4	25	650	4	+39.5	+35.5	..	..	

⁴ Ref. 1, p. 50.

The density change for Fraction 1 as given in the table is that for the sixth run. Runs VII to IX, inclusive, were made on combined light fractions. Run IX gave 55 cc., with $\Delta = -65.2$. Here Δ gives the increase of density in parts per million.

Runs X to XII were made on the basis of a 50% cut. Here the increase of density should be 17.7 parts, and the decrease also 17.7 parts per million. In the tenth run, the 55 cc. from Run IX was split into equal light and heavy fractions, with an estimated $\Delta = -(65.2 + 17) = -82.2$. In the eleventh run this was split in the same way again, with an estimated $\Delta = -(82.2 + 17.7) = -99.9$, and in the twelfth, $\Delta = -(99.9 + 15.9) = -115.8$. Two determinations were made of the density of the 8 cc., or 109 g. thus obtained, and it was found to be -112.4 , which represents an efficiency of 93.3%.

The seventh fractions of Runs I to V were combined, giving 50 cc. with an estimated Δ of $+36$ p. p. m. This was evaporated into 3 nearly equal fractions. The last fraction of 17 cc. was combined with the eighth fractions from Runs II to V, Fraction I₈ having been spoiled by accidental admixture of lower grade material. This 63 cc. with an estimated $\Delta = +64$ was evaporated to 12 cc. with an estimated Δ of $+106.5$, while the value determined is 95.9 p. p. m.

In Runs XX to XXV a cut of 2 was used, and in Runs XXXIX to XLII, cuts of 2 or 3, upon the heavy fractions from previous work. This supplied 16 cc. of mercury with an estimated Δ of $+175$, and a determined Δ equal to 159 p. p. m. This small sample was then evaporated down to 0.2 cc. 7 times, at a rate of 4 cc. per hour, by the use of the small glass evaporator described by Mulliken and Harkins.¹ The combined heavy sample thus obtained, 1.4 cc., was then evaporated to 0.2 cc. with an estimated Δ of $+310$, and a determined value of 260 p. p. m.

Runs XXVI to XXXVIII were made on the combined light fractions, using the steel evaporator in the first 8, and the glass apparatus in the last 5. An intermediate density determination gave $\Delta = -112.5$ p. p. m. instead of the calculated 116. The final 0.4 cc. obtained in this way has a density such that Δ equals 220 p. p. m. instead of an estimated 240.

The total difference of density obtained in this work is equal to 480 parts per million, or 0.096 units of atomic weight, which is practically 0.1 unit. If it be assumed that the atomic weight of ordinary mercury is 200.600, then the heaviest specimen of 3.8 g. has an atomic weight of 200.652, while the lightest, 4.4 g., has an atomic weight equal to 200.556.

Density Determinations

The method and the pycnometers used for the density determinations on *large samples of mercury* have been discussed by Mulliken and Harkins,¹ so only a few examples will be cited here. The ordinary pure mercury used was compared with that of Mulliken.

Density of original material, $\Delta = 0$

Weight of pycnometer and stand
plus mercury (about 101 g.)

1st. detn. = 117.56360

2nd. detn. = 117.56356

3rd. detn. = 117.56357

Mean = 117.56358

Largest deviation from mean

= 2 parts in 10 million

Density of 8 cc. of light fraction

1st. detn. = 117.55233

2nd. detn. = 117.55235

Mean = 117.55234

Deviation from mean = 1 part in 10
million

This gives $\Delta = -\frac{(117.56358 - 117.55234)}{101}$

= -112.4 parts per million

Several slight changes were made in the pycnometer of Mulliken and Har-kins, the principal one of which is the introduction of a small bulb in the upper part of the stem in order to hold the mercury when the temperature in the balance case is higher than that in the thermostat. At the suggestion of Dr. Mulliken the effect of changes of temperature and pressure of the air upon the buoyancy corrections was eliminated by the use of a glass cup of the proper volume, together with that of the glass of the pycnometer. A more accurate method would be to balance the pycnometer with a sealed-up pycnometer of the same area and weight, but this was found unnecessary.

In order to determine the density of fractions as small as 0.2 cc., a pycnometer of about this volume (Fig. 2) was constructed.

Since this differs in certain details from the larger pycnometers, its special features will be described. The capillary is about 0.1 mm. or even less in diameter, and the end of the capillary tube is very carefully ground into the filling device at D. Before using, part of the ground joint on this tube is covered with a thin coat of heavy vacuum grease, care being taken to keep this grease at a considerable distance from the tip, so that none can touch the mercury. The pycnometer is then carefully fitted into the joint at D, the mercury is put into the bulb B, and the filling device is attached to a tube from a mercury condensation pump by means of a greased ground-glass joint which allows the pycnometer and filling device to be rotated in a vertical plane. The apparatus is then evacuated fully while the mercury is in the bulb B, after which it is transferred to C by rotating the apparatus. After evacuating further, the apparatus is suddenly turned so that the mercury drops on the top of the capillary at D. The pycnometer fills completely, and a small amount of mercury is left. The pycnometer is then detached, the grease removed with great care by a solvent which does not spread too easily, and the pycnometer caused to overflow by setting it in a small thermostat at the proper temperature, which was 43.07° with the special pycnometer used. When this is placed in a thermostat at 25.000°, it is found that the mercury comes almost exactly to the mark. Any deviation is determined by a cathetometer or a measuring microscope.

The pycnometer was weighed on a high sensitivity analytical balance, which gave 1 cm. of deflection of a beam of light for 0.14 mg. The balance was made specially sensitive and stable for this work by Ainsworth and Sons.

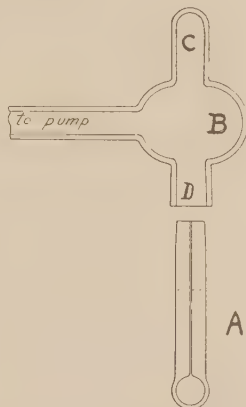


Fig. 2.—Small pycnometer for the determination of the density of mercury (volume = 0.2 cc.).

The weights of the pycnometer and mercury as obtained in one set of determinations are given below.

Weight of small Pycnometer plus		
Light isotopic mercury	Pure ordinary mercury	Heavy isotopic mercury
7.03083	7.03146	7.03217
7.03085	7.03144	7.03213
7.03085	7.03142	7.03211
7.03085		7.03217
Mean=7.030845	Mean=7.03144	Mean=7.032145

While these results are very good, and sufficiently accurate for the purpose, it is believed that the principal cause of the variation which they show is due to the lack of a proper case for the balance, since the error seemed to be due to a variation in the temperature of the balance.

Other density determinations were made but are not listed in this paper, since this would add too greatly to the considerable amount of detail already included. These exhibit the close correspondence between the experimental results and the calculated. While the efficiency in some operations was much lower than in others, the lower efficiency always accompanied some apparent cause, and was usually due to working with smaller quantities of mercury than were required for the most satisfactory results.

Increase of the Density of the Heavy Fraction of an Isotopic Mixture with the Cut

The change in the density with the cut of either the heavy or the light fractions of *any isotopic mixture* may be calculated by the equations of Mulliken and Harkins, provided the composition of the initial mixture (present in the element) is known. The results may be expressed much more simply for the heavy than for the light fractions. It seems to be of value, therefore, to show the forms of the curves which result in the case of some of the most interesting elements. Fig. 3 gives the relative increase of density plotted against the logarithm of the cut. The values for lithium, zinc, and mercury were calculated by the writers, while those for neon and the compounds of chlorine were calculated by Professor A. C. Lunn of the Department of Mathematics of the University of Chicago for one of the writers. He has developed a set of equations which are more exact, but which give results practically indistinguishable from those given by the equations of Mulliken and Harkins.

The curve for zinc is only approximate, since the exact isotopic composition of this element is unknown. It has an abundant isotope of atomic weight 64, and less abundant ones of weights 66, 68, and 70, with a probable isotope of weight 67.

It is of interest that the curves for chlorine and methyl chloride have ordinates which are $\frac{1}{2}$, those for chloroform are $\frac{1}{3}$, and those for carbon tetrachloride $\frac{1}{4}$, of those for hydrogen chloride.

The most important feature of the diagram is that it shows curves of two seemingly different forms, those concave toward the X-axis, as for lithium, boron, etc., and those convex toward this axis, as in the cases of neon and the derivatives of chlorine. The former type of curve occurs

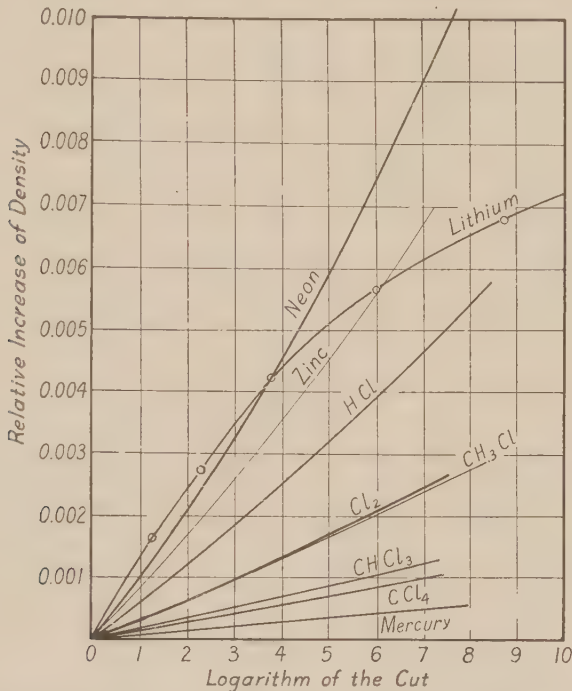


Fig. 3.—Increase in the relative density of various isotopic mixtures as the cut increases. The figure covers all of the region accessible to experiment at the present time, but it is of interest that the upper part of all of the curves is similar to that for lithium, which approaches as a limit the line 0.010. The limits approached by the other curves are much higher, but are all lines parallel to the X-axis.

when the mean atomic weight is nearer that of the heavier isotope, the latter when it is nearer the lighter isotope. It is evident that as the separation proceeds the curves for neon and the derivatives of chlorine will take on the form of that for lithium, and will finally become almost parallel to the X-axis, but at a height dependent on the initial composition of the element. Thus, the complete curves for the different elements have the same general form; but the part of the curve for lithium which is convex toward the X-axis occurs when there is less than 50% of the isotope of atomic weight 7 present, while the natural mixture contains about 94%

of this isotope. Between these two parts of any curve there is obviously a point of inflection. When there are more than 2 isotopes present, there may be several points of inflection.

An apparatus having a capacity of 10 kg. of mercury, with a much increased area, has also been constructed. In this larger apparatus, which has not been used in the work described in this paper, the separation would be much more rapid.⁵

Furthermore, it is obvious that by the adoption of a rectangular instead of a circular cross section of the vacuum jacket, a number (conveniently 12) of troughs of mercury could be arranged side by side, each with its own slanting roof for the condensation of the mercury, and each with its own tube for draining off the fractions. By means of a hole in the roof, glass tubes could be used to supply mercury to each trough. In this way one apparatus would give the same amount of separation as would be produced by 12 units like that described above. On account of the expense of construction, such an apparatus has not been built as yet, but the successful operation of the evaporator described in the present paper demonstrates that the apparatus would operate successfully and rapidly with very little attention if it were provided with mercury seals at all joints, and were evacuated by the use of a rapid pump system.

The writers wish to thank the American Academy of Arts and Sciences for a grant of \$500 from the C. M. Warren Fund. Of this \$300 was spent for a high sensitivity balance, and the rest for apparatus now being used in the separation of the isotopes of mercury, zinc, and cadmium. The separation of mercury is being continued in this Laboratory by Mr. F. Jenkins, who is using the apparatus of 10 kg. capacity as well as those described in the present paper.

Summary

1. By evaporation in a vacuum in a large steel apparatus a separation of the isotopes of mercury has been obtained in which the density difference is 480 parts per million, and the difference of atomic weight is 0.1 unit. This has been secured, without other cooling than that given by ice, in 268 hours of operation of the apparatus, which holds 2.5 kg. of mercury, together with 37 hours of operation of the small glass apparatus used in earlier work in this Laboratory. The weights of the end fractions are 3.8 g. for the heavy, and 4.4 g. for the light. The increase in atomic weight obtained is 0.052, and the decrease 0.044 units. A multiple unit apparatus, which would give the same separation in about 30 hours of operation, has been designed.

⁵ It is evident that the increase in the rapidity of separation secured by this larger apparatus is due to the increased area of the mercury surface rather than to the large volume. The apparatus is so built that the volume capacity of the trough may easily be made small without a reduction of its area.

2. A set of curves has been given to show the relation between the increase in density of an isotopic mixture and the cut. These curves exhibit one curvature when the amount of the heavier isotope is less than 50%, the opposite curvature when the amount is greater, and a point of inflection at this percentage. When several isotopes are present there may be several points of inflection.

A GRAPHICAL STUDY OF THE STABILITY RELATIONS OF
ATOM NUCLEI

By S. L. MADORSKY

Reprinted from a paper in the PHYSICAL REVIEW by W. D. Harkins and S. L. Madorsky

A GRAPHICAL STUDY OF THE STABILITY RELATIONS OF ATOM NUCLEI.

BY WILLIAM D. HARKINS AND S. L. MADORSKY.

SYNOPSIS.

Graphical Study of the Electrical Properties of Atomic Nuclei and their Relation to Stability.—The properties considered are: P the number of positive electrons in the nucleus, which is taken to be numerically equal to the atomic weight, M the net positive charge which is equal to the atomic number, N the number of negative electrons which is equal to $(P - M)$, $[(N/P) - \frac{1}{2}]$ the excess of the relative negativeness N/P over the minimum $\frac{1}{2}$, and n the isotopic number which is equal to $(P - 2M)$ and also to $(N - M)$. The relations between each of these five quantities and each of the others for the various atomic nuclei are shown in ten two-dimensional plots which clearly bring out the stability relations and are of particular interest because of the limited region in each plot where atoms are found. Except in the case of hydrogen and of the helium isotope $P = 3$, N/P is never less than $\frac{1}{2}$ and M/P is never greater than $\frac{1}{2}$; in fact for 85 per cent. of the atoms composing the crust of the earth, both these ratios are equal to $\frac{1}{2}$ and n is equal to zero. As the atomic number, that is the net positive charge increases, the *relative negativeness necessary to stability* increases above $\frac{1}{2}$, more and more; that is, as alpha particles are added extra cementing electrons are required to overcome the increasing mutual repulsion of the positive units of the nucleus. An alpha ray transformation does not change n but increases N/P , while a beta ray change decreases n by 2 units and also decreases N/P ; we therefore find that in each group of isotopes, the ones with larger values of N/P show greater beta-ray and less alpha-ray instability. Stability considerations also help explain the fact that the *number of isotopes* is on the whole smaller for the lighter atoms. The curves bring out other interesting relations. The number of isotopes is larger for even than for odd numbered elements, especially for $M \leq 30$. Also for most atoms, M , n and P are either all even or all odd, but N is usually even. The curve for the *frequency of occurrence of atoms* as a function of n shows periodic maxima four units apart, while as a function of M (or of N) the periodic maxima are two units apart. These regularities make it possible to predict the existence of the more abundant isotopes of elements whose mean atomic weights are accurately known; for instance, in the cases of lithium and boron predictions made in 1920 were later verified by Aston and by Dempster.

IN 1914 Rutherford¹ suggested that the hydrogen nucleus is the positive electron, and that the explanation of the fact that the helium atom has not quite four times the mass of the hydrogen atom, may be due to a "packing" effect. Less than a year later, Harkins and Wilson,² without knowing of this suggestion, published a careful study of the atomic weight relations of the elements, and developed what has since become known as the whole number rule, which is that pure atomic

¹ Rutherford, Phil. Mag., 27, 494-5 (1914).

² Harkins and Wilson, Proc. Nat. Acad. Sci., 1, 276 (1915); J. Am. Chem. Soc., 37, 1367-1421 (1915).

species other than hydrogen, have atomic weights which are whole numbers, or very nearly whole numbers on the basis of oxygen as 16 or carbon as 12. This was stated in the form of an hypothesis: that the packing effect in the formation of helium from hydrogen amounts to about 0.77 per cent., but that the further packing which occurs in the formation of more complex atoms is so slight as to be obscured by the errors in the atomic weight determinations made on pure isotopes. The evidence of the chemically determined atomic weights of pure atomic species seems to justify the statement that while the atomic weight of free hydrogen is about 1.0078, the mean atomic weight of hydrogen in any complex atom is 1.000 ± 0.001 .

Using these ideas as a basis, Harkins has developed a theory of nuclear stability and composition which indicates, among other important relations, that the elements fall into two classes which are on the whole essentially different in both stability and composition. These are: (1) Elements of even number, whose atoms are in general the more stable and the more abundant, and whose nuclei are mostly built up from alpha particles alone or of these together with negative electrons, whenever the nuclear system is not too complicated—that is when the atoms are light. Heavier atoms of this class have in general atomic weights which are divisible by two. (2) Elements of odd atomic number, whose atoms as a whole are less stable and less abundant, and contain in general three (sometimes one or two) hydrogen nuclei in addition to the alpha particles which constitute the greatest part of their composition. It was pointed out that the magnitude of the packing effect is likely to be different for the additional hydrogen nuclei from that found in the alpha particle. In general the atomic weights of pure atomic species of this class are odd numbers. The number of isotopes of elements of even atomic number is in general considerably larger than for elements of odd atomic number.

The atoms may be classified in a different way as (*A*) those of low atomic number (up to 28) which are in general by far the more abundant, and presumably the more stable. The number of isotopes for these elements is in general small. The atoms of elements (*B*) of high atomic number (heavy atoms) are rare, and presumably less stable than those of the light elements, but the number of isotopes (especially for the elements of even atomic number) is much higher.

If the nuclei of atoms are built up from positive and negative electrons, as indicated by the theory, it is easily seen that the number of particles involved is so great, for example 232 positive and 142 negative electrons, or 58 alpha particles, as to make the mathematical treatment of the

stability relations extremely difficult, especially since practically nothing is known concerning the laws of force or of the dimensions or form of the electrons in the nucleus. It seems worth while to make a simple beginning in this direction by plotting graphically the data now known which have a bearing upon the stability of atom nuclei, making use of such variables as seem to be related to this property.

NOTATION.

- P atomic weight, or number of positive electrons.
- M atomic number, or positive charge on nucleus.
- N $P-M$, or number of negative electrons in the nucleus.
- n Isotopic number.
- p a positive electron.
- e a negative electron.
- α an alpha particle.
- pn a neutron.

VARIABLES RELATED TO ATOM NUCLEI.

Two important variables related to the properties of the atom are the atomic weight (P) determined experimentally by chemical methods or by the use of positive rays, and the atomic number (M), which is also determined experimentally. This has been done by the use of the x-ray method of Moseley, and also by the independent method of noting the position of the element in the periodic system of Mendeleeff, which is simply a classification of the elements according to their properties as determined experimentally. The difference of two experimentally determined numbers (N) is also an experimentally determined number. In an exposition of the stability relations two additional dependent variables have been found to be of great convenience. These are the ratio N/P , which may be designated as the *relative negativeness* (presumably of the nucleus), and the isotopic number (n), which specifies the number of an isotope. The isotopic number may be defined as equal to $P - 2M$, so it may also be determined experimentally.

Since the mass and positive charge of an atom seem to be always associated, the term nucleus will be used to refer to that part of the atom which has these properties. The five variables listed above may be assumed to refer specifically to this part of the atom. As only two of the variables are independent, it is obvious that all of them may be represented on a single plane diagram, but, since in such a diagram only two of the axes are found to be at right angles to each other, it is necessary to use rectangular axes for each pair of variables if the relations which

¹ Obviously P/N may be used as the relative positiveness.

exist are to be made sufficiently obvious. In this way ten two-dimensional diagrams are obtained (Figs. 1 to 10).

Of the five variables under discussion only one, N/P , involves the ratio of two others. The other four give rise to six two-dimensional plots (Nos. 1, 2, 3, 8, 9, 10), each of which has the remarkable property that on it *constant values* of every one of the *five* variables, including N/P , are represented by *straight lines*. While this is obviously true of any set of whole number variables defined as these four are, independently of any atomic relationships, the importance of the diagrams consists in the fact that they give a good exposition of the elementary stability relations of complex nuclei. The characteristics of the ten two-dimensional plots are listed in Table I.

TABLE I.

Outline of the Ten Two-Dimensional Plots of the Variables P , M , N , n , and N/P , together with Equations representing Constant Values of the Variables.

(The ratio N/P is represented by R , and $(N/P) - 0.5$ by R' .)

Fig- ure.	Inde- pend- ent Vari- ables.	$R = k$, $R' = k'$.	$N = k$.	$P = k$.	$M = k$.	$n = k$.
1	$\frac{P}{N}$	$N = PR$	$N = k$	$P = k$	$N = P - M$	$N = \frac{P + n}{2}$
2	$\frac{M}{P}$	$P = \frac{2M}{1 - 2R'}$	$P = M + N$	$P = k$	$M = k$	$P = 2M + n$
3	$\frac{M}{N}$	$N = \frac{M(1 + 2R')}{1 - 2R'}$	$N = k$	$N = P - M$	$M = k$	$N = M + n$
4	$\frac{M}{R'}$	$R' = k'$	$R' = \frac{N - M}{2(N + M)}$	$R' = \frac{P + 2M}{2P}$	$M = k$	$R' = \frac{n}{2(n + 2M)}$
5	$\frac{P}{R'}$	$R' = k'$	$R' = \frac{2N - P}{2P}$	$P = k$	$R' = \frac{P - 2M}{2P}$	$R' = \frac{n}{2P}$
6	$\frac{N}{R'}$	$R' = k'$	$N = k$	$R' = \frac{N}{P} - 0.5$	$R' = \frac{N - M}{2(N + M)}$	$R' = \frac{n}{2(2N - n)}$
7	$\frac{n}{R'}$	$R' = k'$	$R' = \frac{n}{2(2N - n)}$	$R' = \frac{n}{2P}$	$R' = \frac{n}{2(2M + n)}$	$n = k$
8	$\frac{M}{n}$	$n = \frac{4R'M}{1 - 2R'}$	$n = N - M$	$n = P - 2M$	$M = k$	$n = k$
9	$\frac{P}{n}$	$n = 2PR'$	$n = 2N - P$	$P = k$	$n = P - 2M$	$n = k$
10	$\frac{N}{n}$	$n = \frac{4R'N}{1 + 2R'}$	$N = k$	$n = 2N - P$	$n = N - M$	$n = k$

Note.—All of the fifty lines for constant values in the ten figures are straight except eight, as can be seen from the equations.

It should be noted that the position of the actinium series in these ten plots is in accord with the relations of this series as they have been given by Soddy; the actual relations being undetermined as yet. According to E. Q. Adams the atomic weight of actinium is an odd number, and that lead from actinium has an atomic weight of 207. This would fit in with the general system fully as well or even better than the scheme proposed by Soddy. Adams' system would be represented if each symbol for an element of this series were to be raised by *one isotopic number*, the atomic number being kept the same.

I. P , N PLOT, AND THE ISOTOPIC NUMBER.

The remarkable feature of the plot which represents the number of negative electrons on the Y axis, and the number of positive electrons on the X axis is shown by the line with a slope of $1/2$ which extends to the right from the origin. In no case is the point which represents the position of any species of complex atoms below this line, though hydrogen, and Rutherford's helium isotope of mass 3 lie below it. However the former of these two is simple, and the latter has not been found to exist as an atomic species. It is of extraordinary interest that 85 per cent. of all of the atoms in the surface of the earth, and 79 per cent. of the atoms in the meteorites are represented by points which lie exactly on this line, on which the relative negativeness to positiveness of the atom nuclei is just as 1 to 2, or $1/2$. Since the formula of any nucleus of this type is $(p_2e)_M$, or $a/2$ it has been suggested by Harkins that the group p_2e may be the most fundamental group concerned in atom building. However, since for more than 99.9 per cent. of the atoms of this formula M is an even number, it seems that, if this group exists, it is very unstable with respect to aggregation, and unites with itself to form groups of the formula $(p_2e)_2$, which are alpha particles.

All of the pure atomic species which are represented by points which lie above this line, lie on 54 lines parallel to it; whose slope is therefore also $1/2$. These are lines of constant isotopic number. The isotopic number of the line of this slope which passes through the origin is zero, the isotopic number of ordinary uranium is 54. The space available for these lines in the diagram is so small that only the zero line is represented. On this plot the lines of constant atomic number have a slope of 1 to 1, or along such lines N must increase by 1 if P increases by 1, so the formulæ of isotopic nuclei differ by one or more neutrons of the formula pe . Thus the nucleus of any atom is represented by the formula $(p_2e)_M(pe)_n$, where n is the *isotopic number*. The chart of the radioactive elements shows, as is evident from the formula of the alpha particle

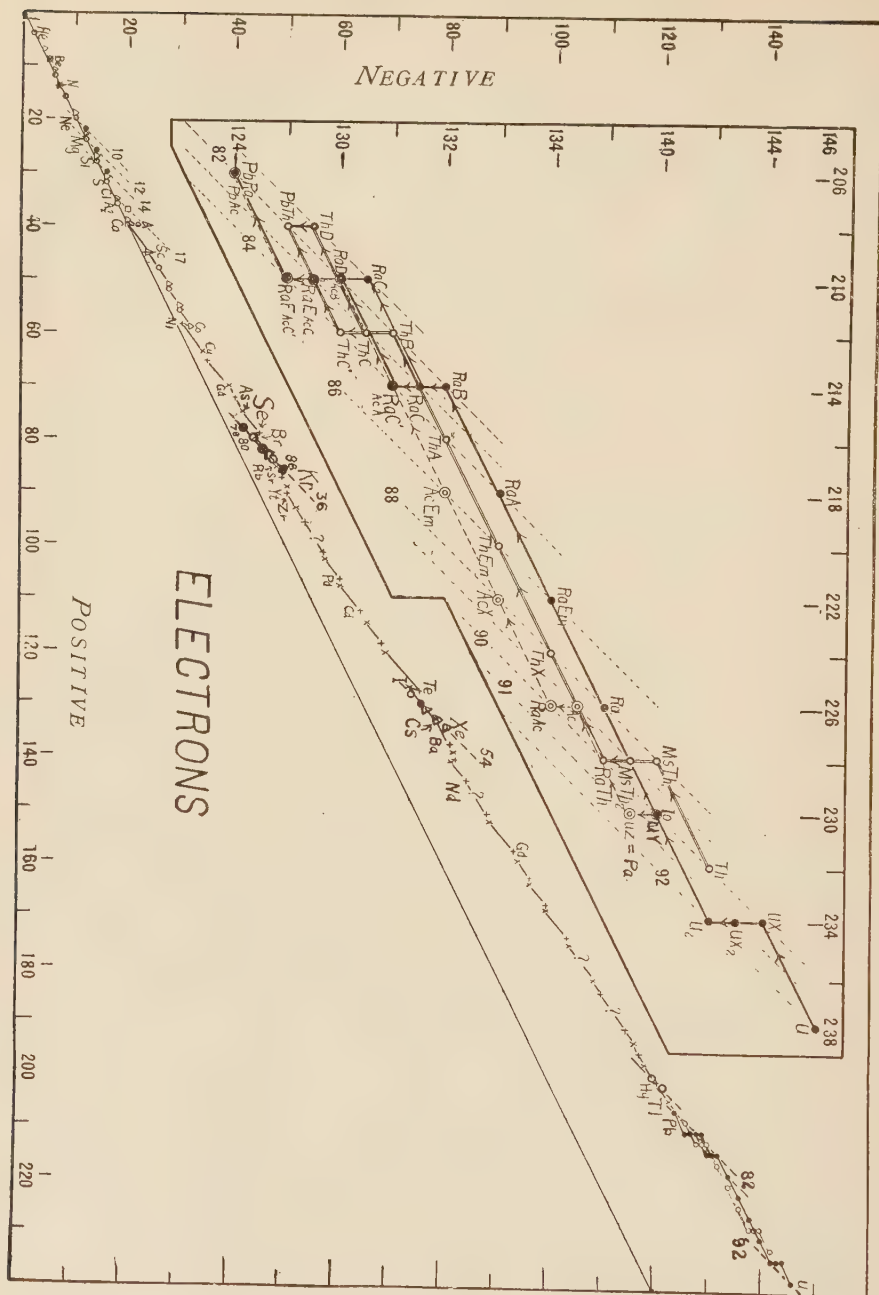
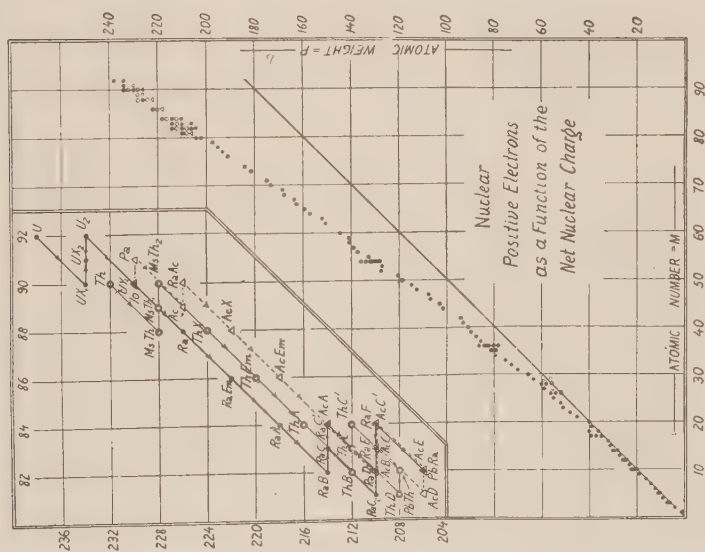
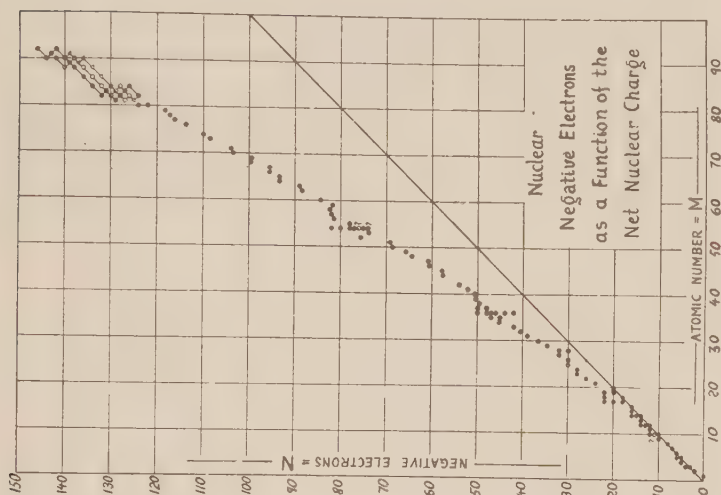


Fig. 1.

Plot of the total number of negative and positive electrons in atom nuclei. Isotopes ($P - N = \text{const.}$) lie on lines (dotted when they are given) which have a slope of 45° . Members of the helium-thorium series are represented by open circles, of the metaneon-uranium series by circles which are inked in, the lithium series by triangles, and metachlorine (37) by a square. Note that the elements of the helium-thorium series from helium to calcium, and the radioactive elements in any series during successive alpha disintegrations, lie on lines which have a slope exactly equal to 0.5.



$(p_2e)_2$, that during a series of alpha disintegrations the isotopic number remains constant. Lines of beta disintegration are lines of constant P , along which the number of positive electrons remains constant. The lines of constant N/P radiate from the origin. Most of the other features of this plot are shown more plainly in some of the other diagrams.

The isotopic number is that number which must be added to twice the atomic number to give the atomic weight, and represents the number of neutrons in the nucleus in excess of the formula $(p_2e)_M$. The value of the isotopic number is given by the following equations developed by Harkins:

$$n = P - 2M, \quad (1)$$

$$n = 2N - P, \quad (2)$$

$$n = N - M. \quad (3)$$

As the atomic number increases the isotopic number also increases in general, or the ratio of N/P increases above its minimum value $1/2$.

2 AND 3. M , P , AND M , N PLOTS.

In the plot on which M is given on the X , and P on the Y axis, the most abundant atomic species lie on a line with a slope of 2 to 1, or in such atoms the atomic weight is twice the atomic number. All other species of complex atoms are represented by points which lie above this line. It is obvious that the ordinates are given by the equation:

$$P = 2M - n.$$

Fig. 3, which gives the atomic number on the X , and the number of nuclear negative electrons, on the Y axis, resembles Fig. 2 very much, the difference being that the line of minimum slope has the slope 1 to 1. Any point on one of these diagrams is the same number of units above the line of minimum slope on one diagram as it is on the other. This number of units gives the isotopic number, or the number of neutrons present in excess of the formula $(p_2e)_M$. The great simplicity of the relations in Figs. 1 to 3 is due to the fact that the minimum value of N/P which is $1/2$, gives the minimum value of P as $2M$, and the minimum value of N as equal to M , since $M + N = P$, or $(M/P) + (N/P) = 1$.

4, 5, AND 6. M , P AND N , ON THE X AXIS; $N/P - 0.5$ ON THE Y AXIS.

Figs. 4, 5, and 6 are very much alike, since the ordinates are the same on all of the three plots. The abscissæ are different, but are very nearly proportional to each other, since M and N are not very different, and P is nearly twice M . Of the three only Fig. 4 will be discussed in detail. In this the *relative negativity* of the nuclei is given on the Y , and the

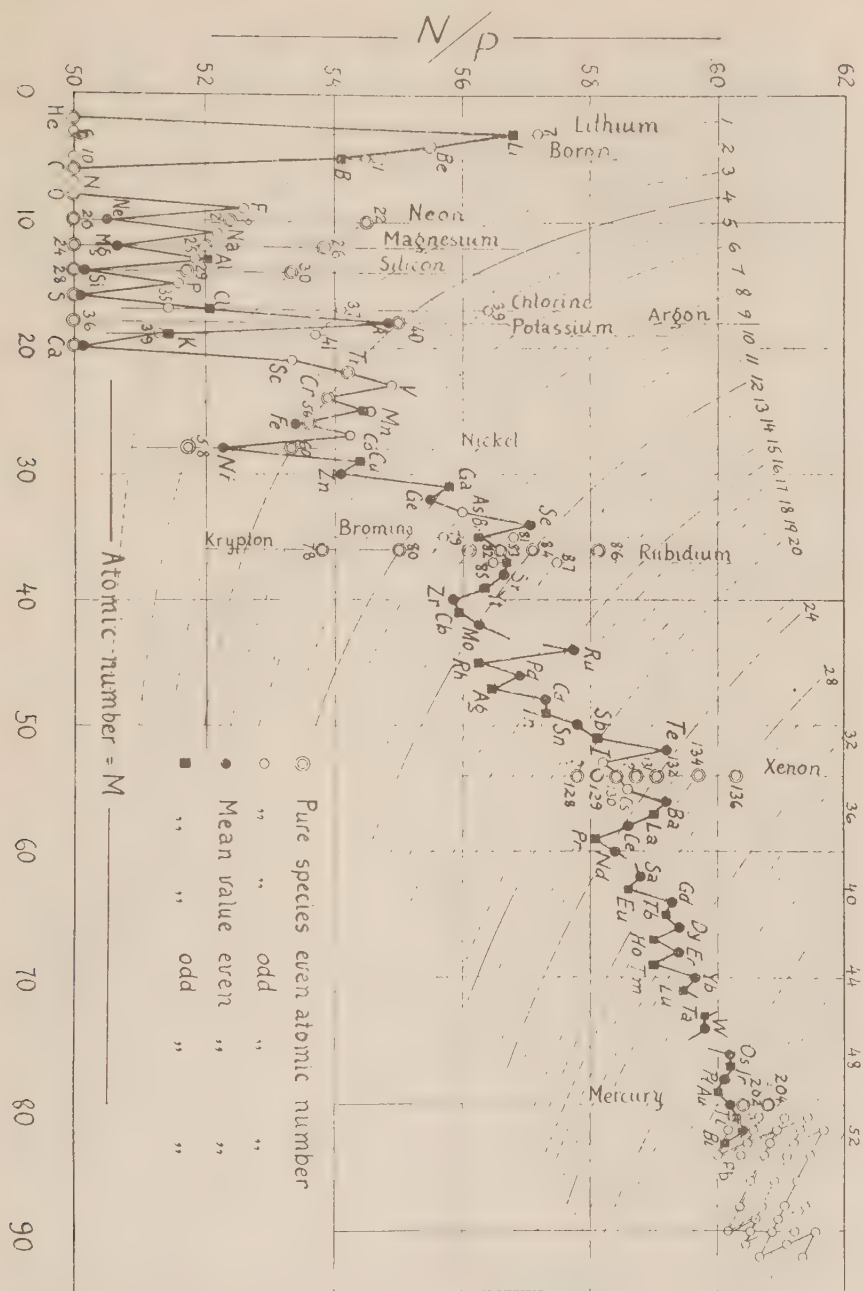


Fig. 4.

The ratio (N/P) of negative to positive electrons in atom nuclei as a function of the atomic number (M). $y = N/P - \frac{1}{2}$; $x = M$.

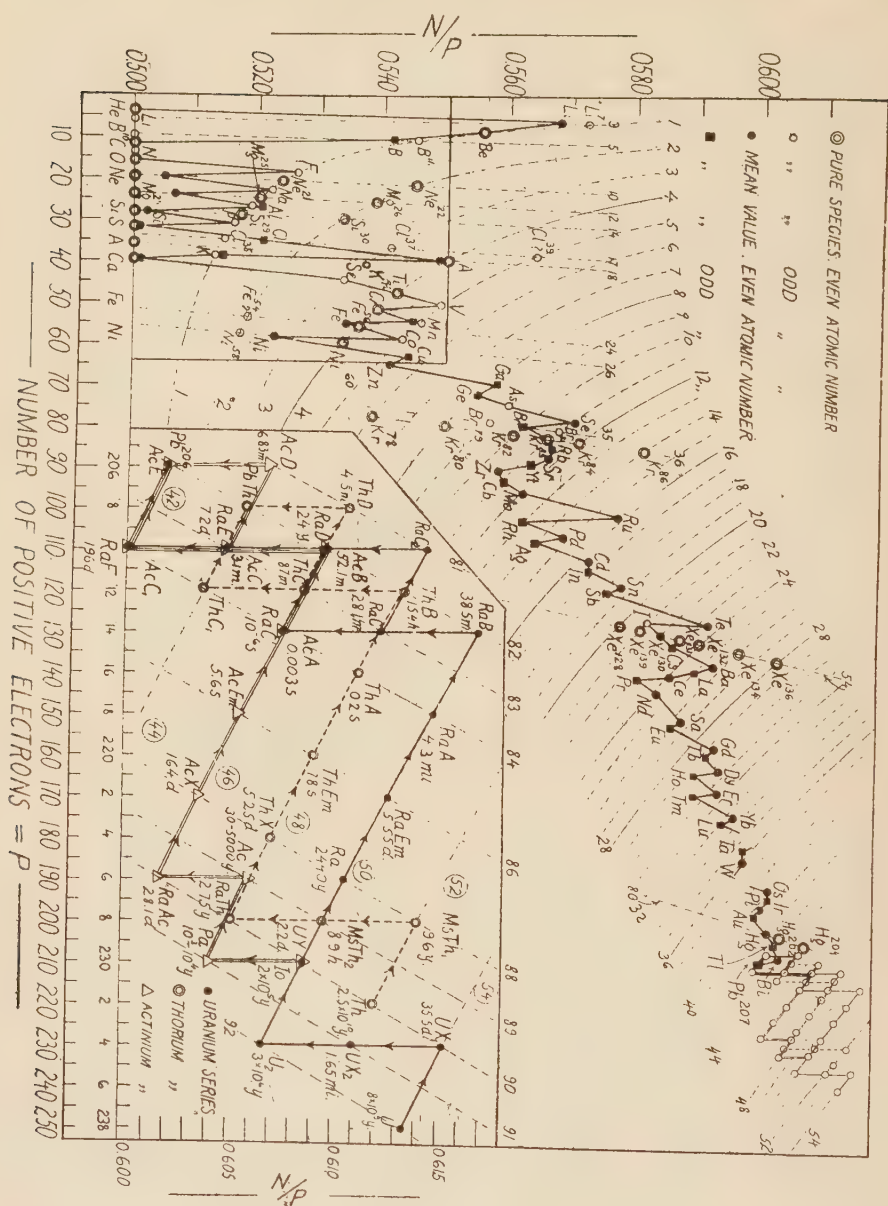


Fig. 5.

Plot showing the ratio of the number of negative to positive nuclear electrons on the Y-axis, and the number of nuclear positive electrons on the X-axis. Atomic species of zero isotopic number lie on the horizontal line $N/P = 0.5$, which is taken as the X-axis of the plot. The isotopic numbers of the other atomic species are given by the numbers of the equilateral hyperbolas.

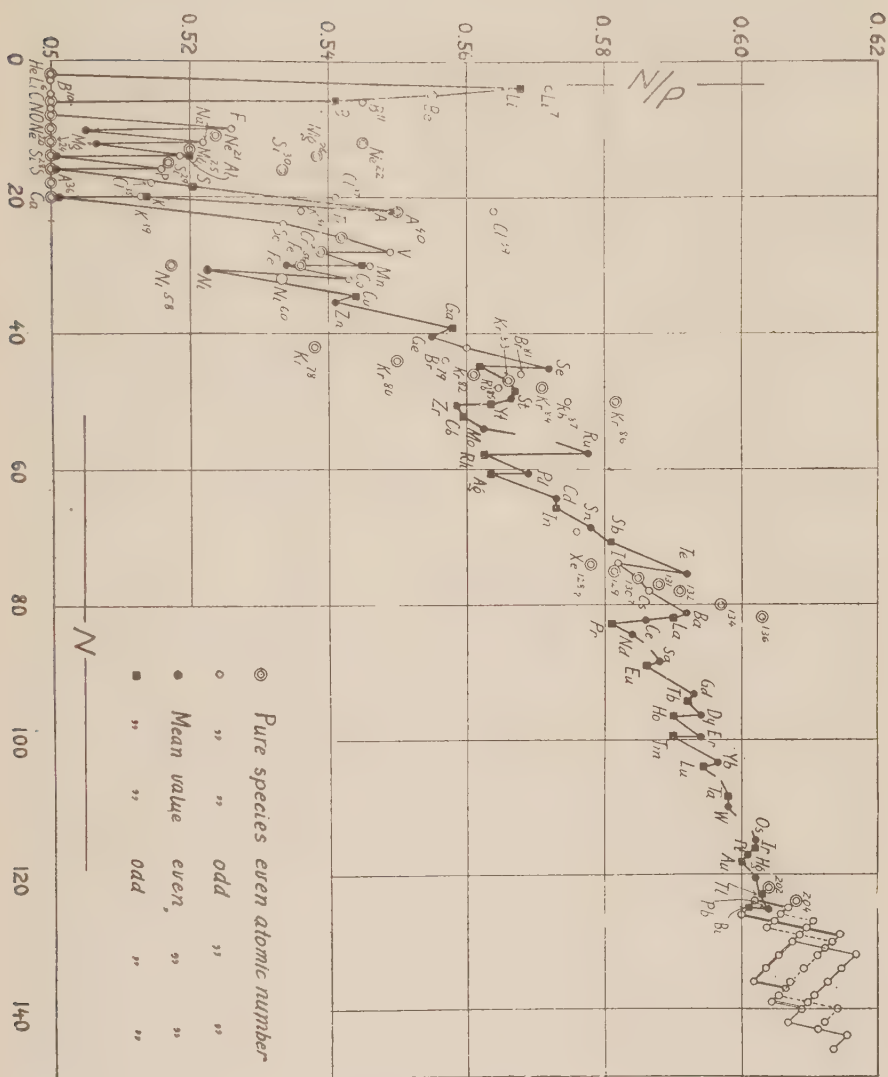


Fig. 6.

Ratio (N/P) of the number of negative to positive electrons in atom nuclei as a function of the number of negative electrons (N).

net positiveness on the X axis. The hypothesis that *the net positive charge on the nucleus exerts a certain self-repulsion, and that this self-repulsion increases with an increase of the charge, provided the relative negativeness of the nucleus remains constant*, has been developed by Harkins.¹ *Stability is secured as the net positiveness increases, by an increase in the relative negativeness.* This is obtained in nuclei built up of alpha particles alone, by the introduction of negative electrons (cementing electrons), used in cementing the alpha particles together. Only cementing electrons are given off in beta disintegrations of the radio-active elements. To a smaller extent an increase in the relative negativeness may be brought about by the addition of groups in which the ratio of N/P is greater than $1/2$. Such groups are p_2e_2 , p_3e_2 , and pe .

Evidence that the relative negativeness (or positiveness) plays an extremely important part in determining stability is found in the relations of the radioactive elements (lower right-hand corner of Fig. 5).² This shows plainly that *if atoms of constant net positiveness* are considered, it is found that as the nucleus becomes more negative in the relative sense, the instability with respect to the emission of negative electrons increases (beta period decreases), and the instability with reference to the emission of positively charged particles decreases (alpha period increases). This indicates that even in the closely packed nucleus, unlike charges attract, and like charges repel, or the sign of the effect is the same as at greater distances. Nicholson and other theorists have assumed, without any evidence for their point of view, that this is not true. When radioactive elements of different atomic numbers are compared, no such relation is found, and it is seen that there is a strong indication that the cementing electrons are arranged in pairs, and the alpha particles in groups of varying number, but averaging about four.

The isotopic lines in Fig. 4 have very nearly the form of rectangular hyperbolas, their equation being $2y(2x + k) = k$, in Fig. 6 their equation is $2y(2x - k) = k$, and in Fig. 5 they are rectangular hyperbolas, $xy = k/2$. In these equations k represents the isotopic number. The stability relations may best be expressed by considering one of these lines, for example that for isotopic number 1. The most abundant elements, aluminium, sodium, and $Mg_{1^{25}}$, lie near the cusp of the curve. In either direction along the curve, as either N/P or M , that is as either the relative negativeness or the net positiveness, increases, the abundance of the atomic species decreases. Going in either direction along any

¹ Harkins, J. Am. Chem. Soc., 42, 1971-3 (1920).

² Fig. 4, if it gave an enlarged diagram of the radioactive elements, would show this even more simply, since the lines of constant atomic number are vertical, but the two plots are so nearly alike that it seems unnecessary to duplicate this part of the figure.

one of these lines, starting at the position of maximum stability of the nucleus, the instability of the atoms decreases so greatly that after a certain, not very great distance is traversed, no atom is capable of existence for any appreciable time.

It has been shown by Harkins that the nuclear properties are to a considerable extent periodic, the periodicity being between elements of even and odd atomic number. Figs. 4, 5, and 6, indicate that among the light elements (atomic numbers 2 to 32) the mean value of the *relative negativeness* is much higher for elements of odd than for those of even atomic number (except in the case of three transition elements, Be, A, and Sc). Curiously enough, among the elements of higher atomic number, the relative negativeness is in general considerably higher for the elements of even atomic number, since all of the high peaks, and most of the minor ones as well, represent elements of even atomic number. In all of these three figures the lines of constant atomic weight (P) slope more to the left than those for constant atomic number (M), while the lines representing a constant number of negative electrons, slope still more to the left. This is shown by the equations given in Table I.

7. n , $(N/P) - 0.5$ PLOT.

Fig. 7 illustrates the increase of the relative negativeness of the nuclei as the isotopic number increases. All of the extremely abundant atomic species, with the exception of iron and aluminium, are represented by the *point* N/P equal to 0.5, with an isotopic number zero. The region of highest stability has very nearly the form of a parabola, and along any isotopic line the stability decreases on passing vertically higher or lower. As an example the lines showing the net nuclear charge as 92, the number of positive electrons as 238, and the number of negative electrons as 146, which are the values for ordinary uranium, are given. These lines indicate that for a nucleus with any of these three properties, represented by relatively large numbers, to exist, the isotopic number must increase to a sufficiently high value (about 54) to cause the value of the relative negativeness to become sufficiently high (about 0.614) that the repulsion due to large net positiveness may be overcome by the high relative negativeness.

The diagram in the lower right-hand corner of Fig. 7 indicates that during alpha disintegrations the isotopic number remains constant, while the relative negativeness (N/P) increases slightly. After several changes the nucleus becomes so negative in the relative sense that positively charged particles no longer leave it, but negative electrons are shot off. Each beta disintegration lowers the isotopic number by

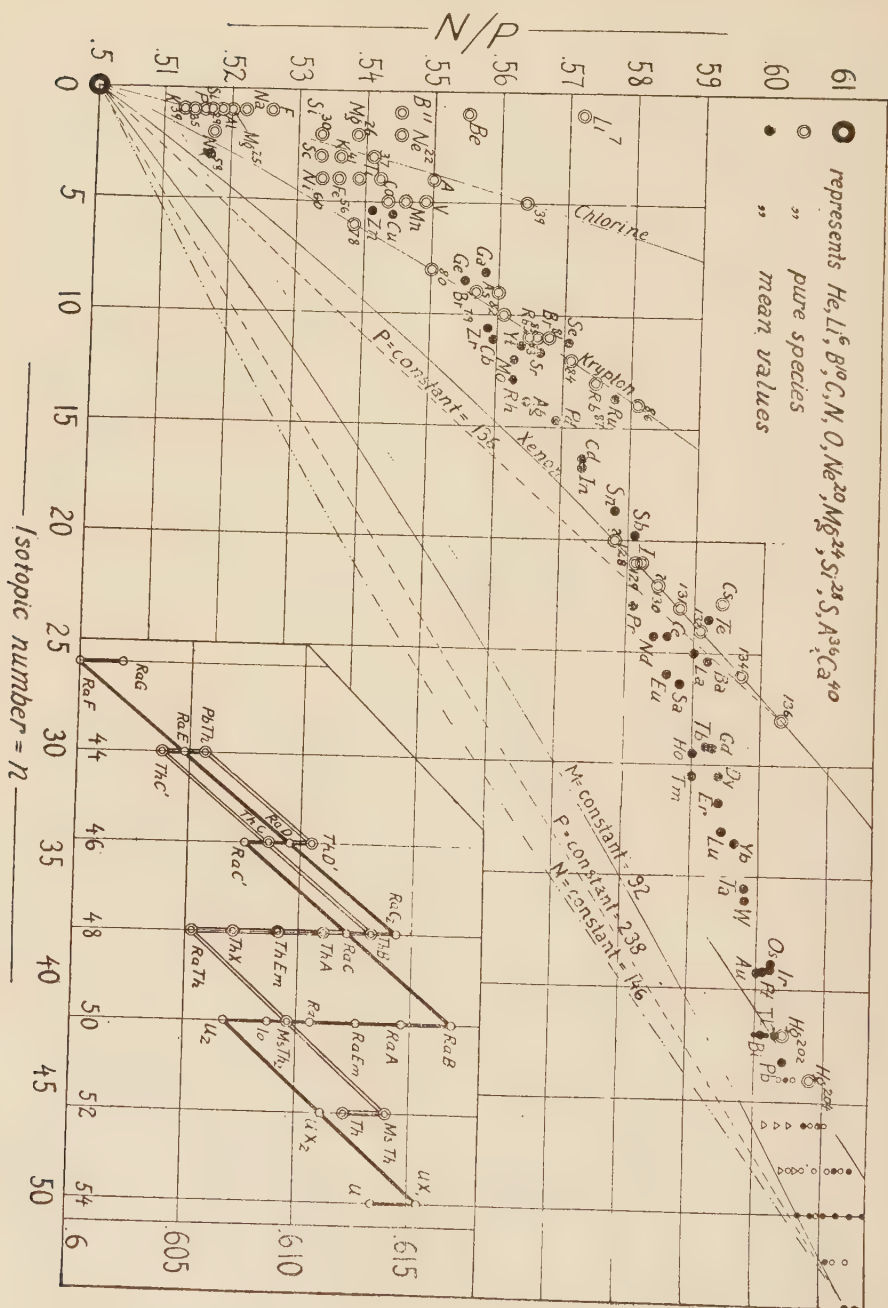


Fig. 7.

Ratio (N/P) of the number of negative to positive electrons in atom nuclei as a function of the isotopic number (n).



On this plot constant values for (1) the net positive charge on the nucleus, M ; (2) the number of hydrogen nuclei or positive electrons in the nucleus, P ; (3) the number of nuclear negative electrons, N ; (4) the isotopic number, n ; and (5) the ratio N/P , are all represented by straight lines.

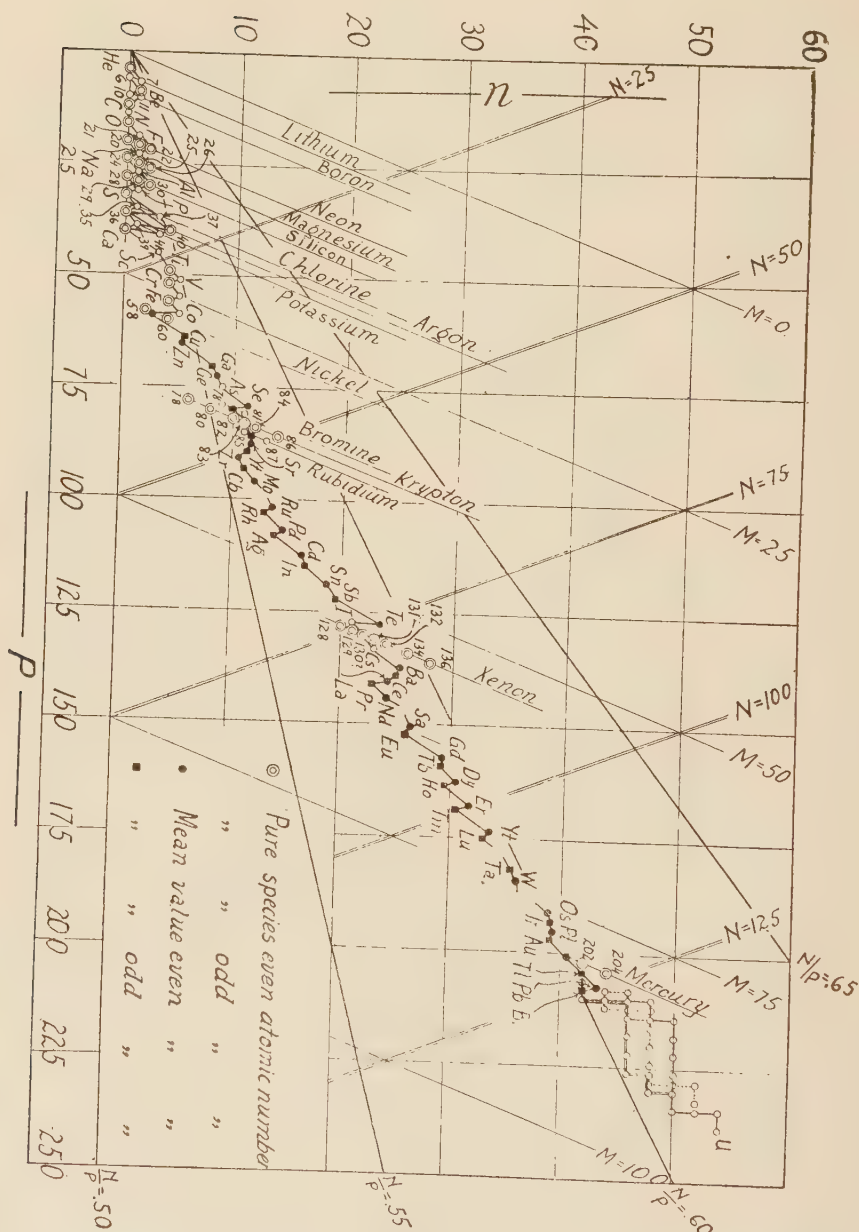


Fig. 9.

Isotopic number as a function of the number of positive electrons.

two, and in this region decreases the relative negativeness slightly more than two alpha disintegrations. It may seem strange that the loss of one negative charge changes the numerical value of the ratio N/P more than two losses of doubly charged positive particles, but the reason for this is obvious, since the loss of an alpha particle decreases both P and N . It is apparent that beginning with uranium, 27 beta changes would be sufficient to bring the plot to the origin, independently of the number of alpha changes, so this is the highest number of beta changes of this type possible for any known atom.

8, 9, AND 10. M , P , OR N ON THE X AXIS, n ON THE Y AXIS.

Figs. 8, 9, and 10 represent the isotopic number on the Y axis. All of them have the property that constant values of all of the five variables are represented by straight lines; four, P , M , N , and n , by sets of parallel straight lines, and N/P by lines radiating from the origin. The heavy lines in the radioactive region indicate the paths of the decompositions, but in the remainder of the diagram are only intended to bring out the position of the region of highest stability, which takes on very nearly the form of an hyperbola. A part of this figure has been considerably enlarged, and is shown in Fig. 11, which will serve to indicate the relations more clearly.

In figure 11 the lines for four of the variables, P , N , M , and n , all meet at every corner. Each such corner corresponds to the existence of a *possible* isotope, but not every corner corresponds to a condition of stability. From the general theory already presented it is to be expected that most of the corners which are filled will represent an *even number* of negative electrons. This means that when the atomic number (M) is even, the isotopic number (n) is also even, and when the atomic number is odd, the isotopic number is also odd. *Thus in general the atomic number, isotopic number, and number of positive electrons, all match in the sense of oddness or evenness*, that is all of these numbers are either even, or else they are all odd. In the range of this figure, 3 out of the 31 atomic species thus far found, do not follow this rule. The secondary relation here revealed is that in a small fraction of the known atomic species P and N are both odd together. Only in extremely exceptional cases (less abundant isotopes of Li and B) is N odd, and P even.

Figure 11 has exactly the characteristics which it would have if across it had been plotted the tracks of four radioactive disintegration series, and it gives ample evidence that the light atoms from carbon to zinc as well as the heavy atoms are built up from alpha particles of mass 4, for along each isotopic line the atomic number jumps by 2 and the atomic weight

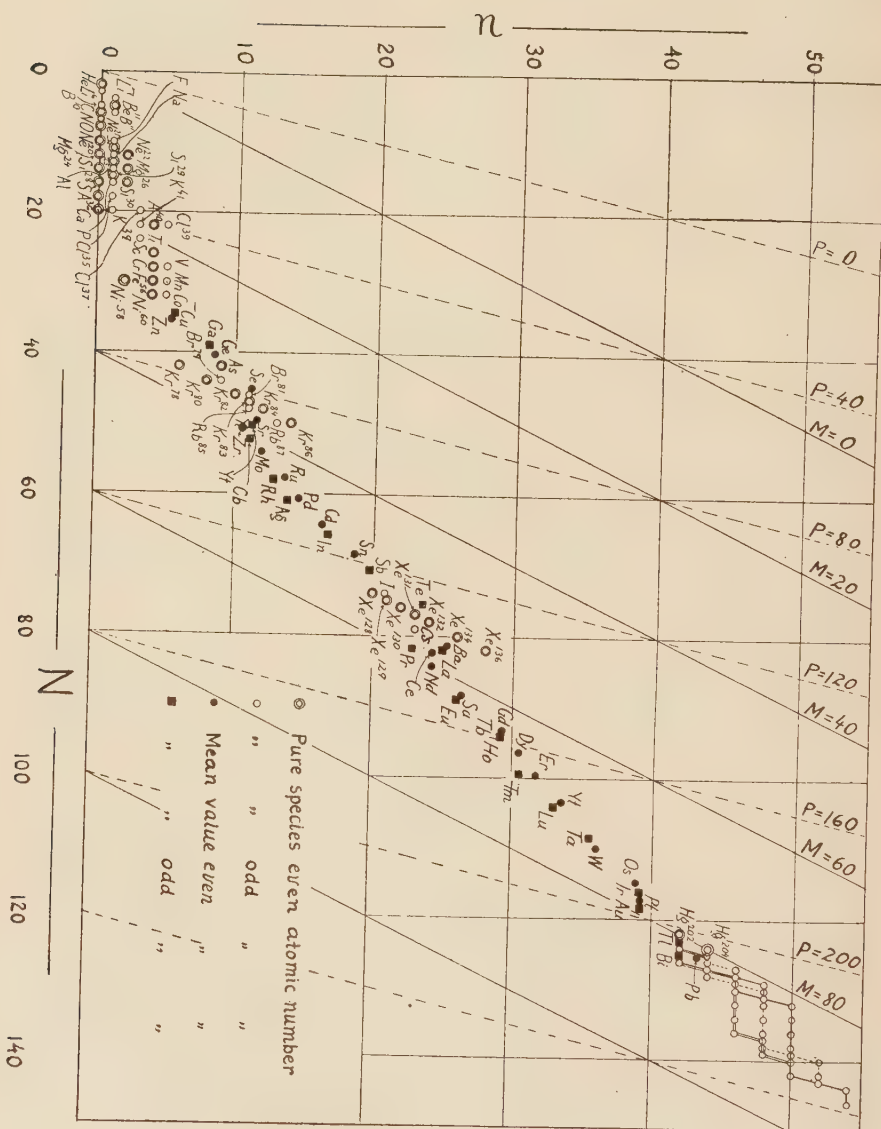


Fig. 10.

Isotopic number (n) as a function of the number of negative electrons (N).

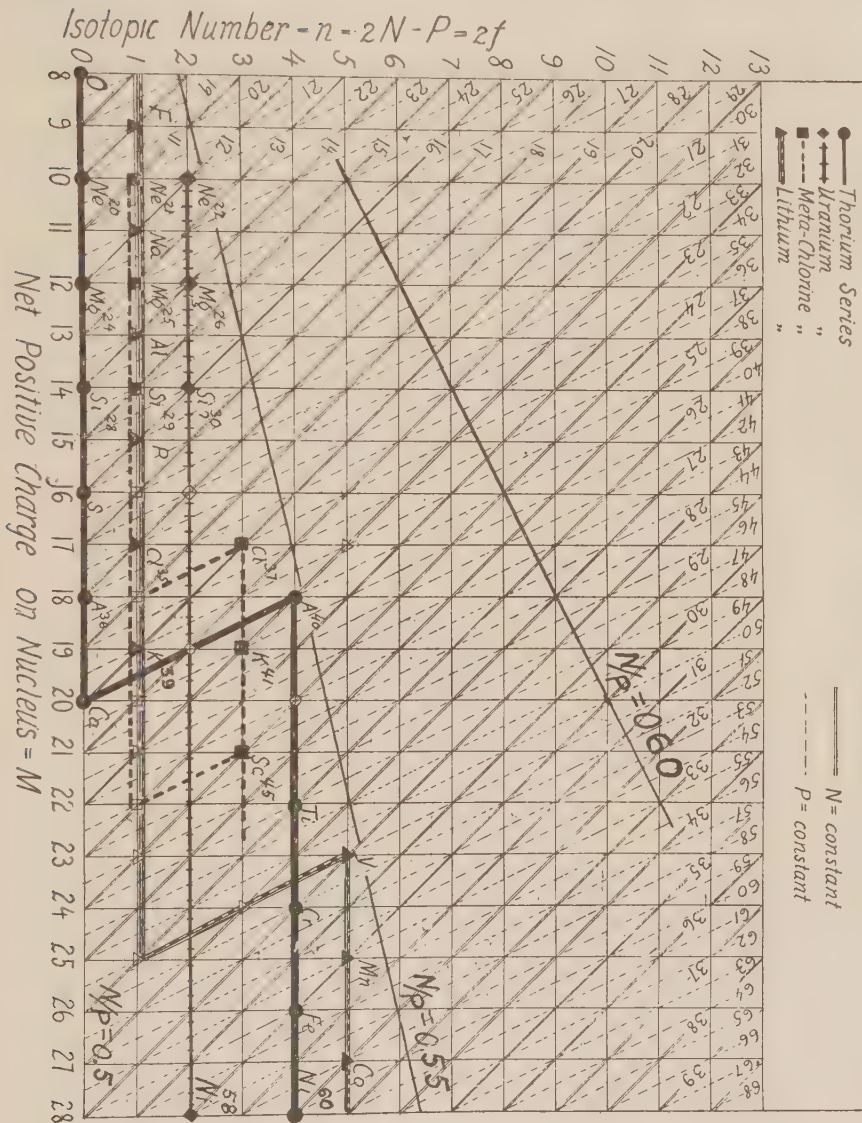


Fig. 11.

(Enlarged section of Fig. 8.) Isotopic number as a function of the net nuclear charge (M)

by 4. Only between fluorine and phosphorus do two of these series become intermingled. Out of the 14 atomic species depicted as belonging to the Helium-Thorium Series, only two remain undiscovered. One of these, K^{41} , probably does not exist in quantities large enough to be easily discovered. The other is Ca^{44} . The form of such a plot is determined by the stability relations of the atoms. The figures of this paper suggest that the light atoms, as well as the heavy ones, both build up, and disintegrate. The ratio N/P changes much more markedly with the isotopic number among the light than among the heavy atoms, so the range of the isotopes in terms of the isotopic number or atomic weight, is much less when the atomic number is low. It is of interest to note that in Fig. 11, while the value 0.55 for the value of N/P is practically attained three times, in no place is it exceeded. While in this range atoms with values of N/P higher than this limit may be discovered, it is now apparent from the atomic weights and the general relations involved, that no such atomic species will be at all abundant.

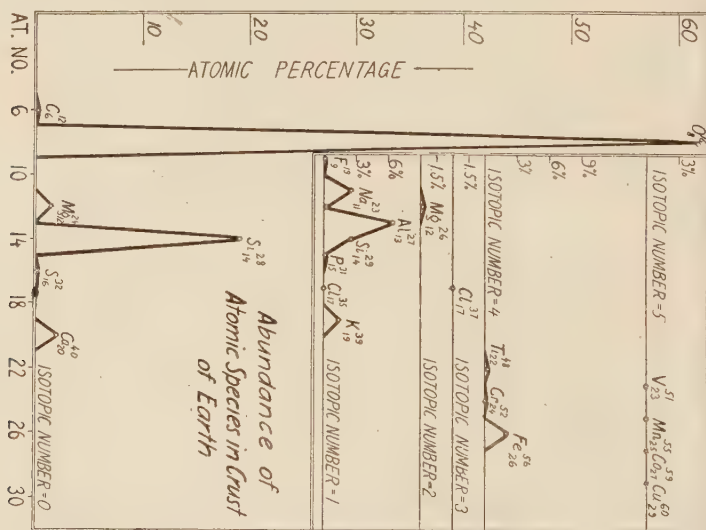


Fig. 12.

Shows the periodic variation in the abundance of the atomic species of each isotopic number.

That the abundance of the atoms of any isotopic number is periodic, is shown by Fig. 12. The abundance is greatest for isotopic number 0, and decreases gradually to a minimum in isotopic number 3, rises to a secondary maximum in isotopic number 4, and again decreases. Thus there is also a periodic variation of the abundance of the atoms in the isotopic numbers, which gives a maximum with each fourth isotopic

number. While these maxima probably become much less prominent as the atomic number increases, their prominence in the range of low atomic numbers is strongly suggestive that the periodicity by 4 is due to the fact that 4 is the mass of the alpha particle.

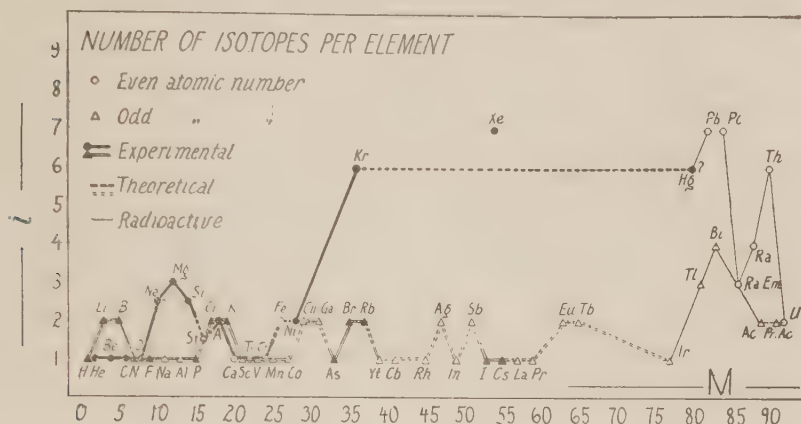


Fig. 13.

Approximate number (i) of isotopes per element.¹

¹ This figure plots the number of isotopes per element thus far discovered, using inked-in symbols. For elements whose isotopes have not been determined a rough estimate of the probable number is given. This number is based upon the assumption that the chemical atomic weight is entirely exact. In so far as this assumption is not justified, the number of isotopes predicted cannot be expected to be correct; and it is well known that many of the chemical atomic weights are very much in error. This is illustrated by the case of antimony, the atomic weight of which has been changed more than a unit by the most recent determinations. The number of isotopes in neon and silicon is plotted as 2.5, an impossible number, to indicate that Aston's experimental determinations are indefinite, giving in each case two isotopes, with a third doubtful. While the number of isotopes plotted for the radioactive elements is greater than for the preceding elements, this is only because the experimental means used for their detection is much more delicate. While Aston was able to find only one atomic species for sulphur, the chemical atomic weight indicates that sulphur consists of at least two isotopes. To show the relations properly the plot should be three-dimensional, and should give the abundance of each isotope. For example, in the two-dimensional diagram lithium, with only 6 per cent. of the higher isotope, is given the same rating as chlorine with 23 per cent. and bromine with about 50 per cent., which gives a false impression unless these facts are kept in mind. The designation of indium, columbium, yttrium, lanthanum, and praeaseodymium as consisting of only one isotope each, is probably without meaning, since in most of these the atomic weight is not known with sufficient accuracy as to give a basis for a definite prediction. On the other hand the atomic weights of silver and copper are known with sufficient precision as to justify the prediction that each of these elements consists of two isotopes. It is of course possible that three isotopes may exist, but the number of isotopes sufficiently abundant to be detected by the positive ray method in its present degree of sensitiveness, is in general not greater than two for elements of odd atomic number.

The most important feature of this plot is that it illustrates the fact that the number of isotopes is in general much higher for elements of even than for elements of odd atomic number, especially for elements of atomic numbers greater than 30. This is exactly in accord with the

Figure 13 shows the way in which the number of isotopes per element varies with the atomic number. It will be seen that the relations are exhibited most easily when separate curves are drawn for the elements of even and of odd atomic numbers. The lines represent only the most general form and not the details of the numerical relations.

While Aston did not find a higher isotope of sulphur, there is little doubt of the existence of such an isotope, which would have an atomic weight of either 34 or 33. Also there is little doubt that a higher isotope of calcium (possibly 42 or 44 atomic weight) and a lower isotope of iron (probably 54) exist. The stability relations contain specific factors of such importance that it is impossible to tell exactly the relative abundance of such isotopes. However it may be pointed out that Harkins predicted the existence of the 6 and 7 isotopes of lithium, the 10 and 11 isotopes of boron, and the 24 and 26 isotopes of magnesium without making a single error in the predictions, but failed to predict the existence of the 25 isotope of magnesium, for which the relations are much more specific.

UNIVERSITY OF CHICAGO,

September 12, 1921.

predictions from the theory of the stability of atom nuclei as presented by Harkins in various papers published in the years 1915 to 1917. (It is of interest to note that the number of isotopes of element 30 (zinc) has just been found to be 4 by Dempster. Thus the number of isotopes increases exactly at the point predicted by Harkins in the year 1915.)

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Published Monthly

Two Volumes Annually

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